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NITROGEN INFUSION R&D FOR CW OPERATION AT DESY

M. Wenskat[1]*, C. Bate[1,2], T. F. Keller[1], A. Dangwal Pandey[1], B. Foster[1,2,3], D. Reschke[1], J. Schaffran[1], G. Dalla Lana Semione[1,2], S. Sievers[1], A. Stierle[1,2], N. Walker[1], H. Weise[1]
[1] Deutsches Elektronen-Synchrotron, 22607 Hamburg, Germany
[2] University of Hamburg, 20148 Hamburg, Germany
[3] Physics Dept., University of Oxford, Oxford, UK

Abstract

The European XFEL cw upgrade requires cavities with reduced surface resistance (high Q-values) for high duty cycle while maintaining high accelerating gradient for short-pulse operation. To improve on European XFEL performance, a recently discovered treatment is investigated: the so-called *nitrogen infusion*. The recent test results of the cavity-based R&D and the progress of the relevant infrastructure is presented. The aim of this approach is to establish a stable, reproducible recipe and to identify all key parameters for the process. Advanced surface analysis is carried out on cut-outs of cavities and samples treated together with cavities. Techniques used include SEM/EDX, TEM, XPS, XRR, GIXRD and TOF-SIMS. The aim of this approach is to establish a stable, reproducible recipe, to identify key parameters in the process and to understand the underlying processes of the material evolution, that result in the improved performance observed.

THE EUROPEAN XFEL

The successful construction of the 17.5 GeV European X-ray Free Electron Laser (XFEL) [1] represents the culmination of over 20 years of superconducting radio-frequency (SRF) R&D. The core high-gradient 1.3 GHz nine-cell niobium cavity and cryomodule technology was originally developed by an international collaboration coordinated by DESY for the TESLA (TeV-Energy Superconducting Linear Accelerator) project — a 500 GeV center-of-mass electron-positron linear collider and integrated X-ray FEL, proposed in 2003 [2].

A total of 102 cryomodules containing 816 cavities have been produced, of which 97 have been installed in the SRF linacs of the European XFEL, which represents by far the largest deployment of TESLA technology to date [3,4]. The cavities achieved an average usable accelerating field of 29.7 ± 5.1 MV/m and an average quality factor Q_0 of 1.2×10^{10} at the design field of 23.6 MV/m in the vertical acceptance test. The design energy of 17.5 GeV of the superconducting linac was met beginning of August 2018.

The design beam pattern for the user operation of the European XFEL has a duty factor of 1.4% with 2.7k bunches per second accelerated up to the maximum energy. As a future upgrade, more flexible beam patterns are planned, allowing 50k bunches per second and a duty factor of 10-50% up to 10 GeV or a continuous wave (cw) mode with 100k bunches per second up to 8 GeV are planned. In order to achieve these duty factors, higher quality factors of the cavities at medium gradients are necessary.

The current plan for the upgrade is to exchange the first 17 cryomodules (136 cavities) in the tunnel (injector plus section L1 and L2) and possibly lengthen L3 using the cryomodules removed which is otherwise kept unchanged, see Fig. 1. An installation of a SRF gun capable of cw operation, and one Klystrode (IOT) for each rf station of 4 modules is further necessary. This will allow the operation of the European XFEL with short pulses at high energies or with high duty cycles at moderate or low energies [5]. Two R&D topics have been identified to increase the Q_0 of the cavities: *large-grain* (lg) material and *nitrogen infusion* [6,7].

Large-grain niobium material has been studied during the European XFEL preparation phase and is characterized by an average grain size on the order of centimeters, which is several orders of magnitude larger than the standard grain size of ≈ 50 μm of the material (fine-grain or fg) for the European XFEL cavity production. Eleven nine-cell and several three- and single-cell cavities have been fabricated and showed on average a 25% higher quality-factor Q_0 compared to fine-grain material with the same surface treatment. Furthermore, a pre-series cryomodule called XM-3, consisting of seven lg and one fg cavities, shows excellent results in cw operation [8]. Recent module tests showed stable operation at 17 MV/m with a Q_0 of 2.3×10^{10} at 2K and long pulse operation with duty factors from 22 - 43% up to 22 MV/m.

The emphasis of this paper will be on the investigation and application of a process called *nitrogen infusion*, which was first presented by Fermilab [7]. Several laboratories including DESY are working on the understanding of the process and its influence on the cavity surface and the relation of the changed surface to the rf performance [9–11]. The current status and plans of DESY's efforts are presented here.

INFUSION R&D AT DESY

The R&D program at DESY has three approaches:

- treating cavities in a large furnace, together with samples as quality control, to reproduce the performance improvements
- treating samples in a UHV mobile chamber with in-situ surface characterization to observe and understand the surface evolution

* marc.wenskat@desy.de

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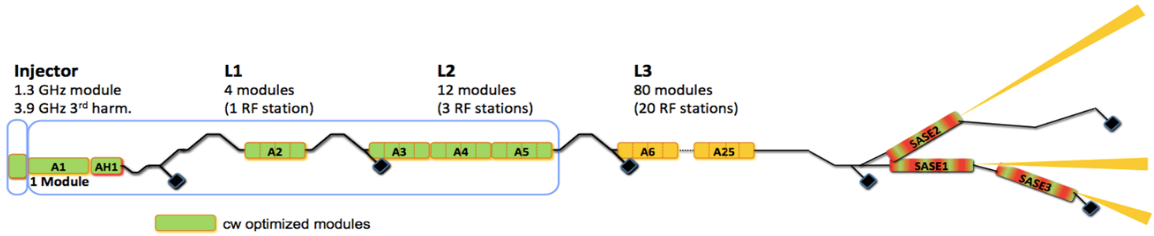

Figure 1: The first 17 modules (136 cavities) of the European XFEL need to be replaced to achieve the desired flexibility for the user operation. An SRF gun based cw injector and one IOT for each rf station is necessary. L3 remains unchanged.

- treating samples in a small furnace to scan the parameter space and optimize the infusion recipe

In addition, a collaboration with other laboratories regarding the exchange of experience, data, cavities and samples has been established or is in the process of being established.

Infusion Recipe

The actual infusion recipe includes a heat treatment of 800°C for 3 hours in a pressure below $5 \cdot 10^{-6}$ mbar. It is widely regarded to be necessary to partially break up the natural surface oxides and create a reactive Nb surface. While still in vacuum, the cavity is cooled down to 120°C and kept there for 48 hours. Nitrogen is injected at a partial pressure of 0.03 mbar which is actively maintained. During this phase, it is assumed that nitrogen enters the material as an interstitial. After 48h, the furnace is evacuated and the cavity is cooled down to room temperature. An additional point is the installation of so-called *caps* onto a cavity before the treatment, see Fig. 2. Initially, they were installed to prevent a Ti contamination in the FNAL furnace from entering the cavity. How these caps affect the inner atmosphere during this procedure, act as a filter or getter material or if they are necessary at all has not yet been clarified.

Cavity and Sample Preparation

Cavities underwent a standard reference test at 2K before any further preparation was done. If a cavity showed a good performance, e.g. similar to the blue data points in Fig. 3, it was prepared for the infusion procedure. This preparation includes the disassembly of all antenna and vacuum components, a high-pressure rinsing of the inner surface to remove any particle contamination from the disassembly, and the installation of the Nb caps onto the flanges. The Nb caps underwent a chemical polishing of ≈ 10 µm before use. Afterwards, the cavity was sealed in a clean-room foil bag and transported to the furnace. The bag was opened under local clean-room conditions and the cavity installed into the furnace.

The samples used are made of standard European XFEL material. Sheets from three different vendors which were still available after the cavity production were cut in a series of standardized sample shapes. The samples underwent the standard European XFEL cavity preparation, including electrochemical surface removal of ≈ 120 – 140 µm, a first outgassing bake at 800°C for 3 h, a second chemical polishing of ≈ 30 µm and another bake of 120°C for 48h. This recipe has been developed over years, see e.g. [12] for details. The samples are placed within an Nb box made from cavity-grade material to simulate the interior of the cavity.

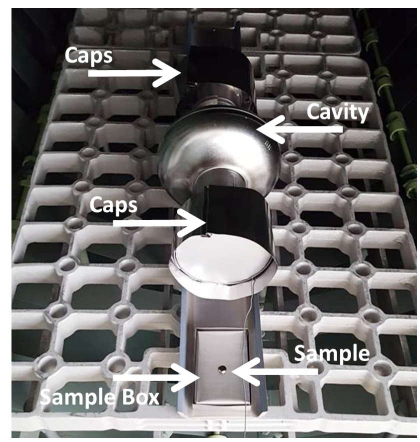

Figure 2: 1DE27 installed in the furnace and the temperature sensor is attached to it. Samples are placed inside a Nb box to simulate the interior of a cavity. One sample for comparison is placed outside the box. The caps cover the flanges of the cavity.

Cavity Furnace

The furnace in use has a volume of 7 m^3, of which an effective volume of $1800 \times 625 \times 660$ mm^3 has a homogenous temperature during baking, and was produced by the company Ipsen. The maximum achievable, stable temperature is T_{max} = 1100°C and the furnace has a temperature stability of ±2°C. The door of the furnace is double sealed by EPDM O-rings with a vacuum in between. Other accessories such as pumps, gauges etc. are sealed by standard EPDM seals. The pre-pumps are rotary vane pumps and a roots pump. The main pumps are two Varian turbo-molecular pumps (TMP) with a pumping speed of 6000 l/s each. The pressure during the baking is monitored with an ionization gauge IKR050 by Pfeiffer Vacuum which is sensitive between $2 \cdot 10^{-9}$ mbar and $5 \cdot 10^{-3}$ mbar. Residual Gas Analysis (RGA) are taken during the 800°C bake and before and after the nitrogen injection at 120°C. Prior to each infusion run, a bake out at T_{max} = 1100°C for 3 h is done. The installation in the fur-

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nace of a single-cell cavity with niobium caps, the niobium samples in a Nb box can be seen in Fig 2.

RESULTS

Cavity Tests

A total of six cavities have been treated within the R&D program so far. The first cavity, a single-cell cavity, 1DE18 was treated but the rf performance deteriorated after the treatment. During the infusion run, the nitrogen pressure was one to three order of magnitudes lower than the FNAL recipe specified. The reason was that the mass flow was controlled at the nitrogen injection line while the TMP was fully operational, reducing the effective nitrogen pressure in the furnace. As a check of the furnace and to see if a failed nitrogen injection or a pollution of the nitrogen was the cause of the deterioration, a second single-cell cavity 1DE17 was treated similar to 1D18 but without an injection of nitrogen. The cavity deteriorated in the same way. A third cavity, 1DE16 was tested and showed the same characteristics. The key bake parameters and the rf test results before and after the treatment are given in Table 1 and Fig. 3.

Table 1: RF Results and Key Bake Parameters of the First Three Cavities

	1DE18	1DE17	1DE16
Material	Ningxia fine grain	Ningxia fine grain	Plansee fine grain
Reference @ 2K			
$E_{acc,max}[\frac{MV}{m}]$	37.7 - BD	31.2 - BD	32.2 - BD
$Q_0(4\,MV/m)[\times 10^{10}]$	2.8	2.5	2.7
Baking Parameters			
p @ 800°C [mbar]	2×10^{-5}	1.1×10^{-5}	5.5×10^{-6}
P_{N_2} @ 120°C[mbar]	$7-300 \times 10^{-5}$	w/o	w/o
RF Test @ 2K			
$E_{acc,max}[\frac{MV}{m}]$	20.2 no FE	19.5 no FE	26.3 - BD no FE
$Q_0(4\,MV/m)[\times 10^{10}]$	0.5	1.2	3.2

A detailed analysis of the RGAs obtained during these runs showed rather large contributions from hydrocarbons. Samples from all runs showed C-enriched precipitates on the surface - see e.g. the SEM image in Fig. 4.

Improvements to the pumping systems were carried out with the aim of reducing uncertainties and possible back-flow from the pre-pumps. A subsequent test run of a single-cell cavity, 1DE9, was then carried out. The base pressure at 800°C was 5×10^{-6} mbar. The performance deteriorated again, but showed a different behavior, see Fig. 5.

This behavior was observed before at KEK in their failed infusion runs [11]. A comparison of samples from KEK runs is currently being made, to look for common features. An analysis of all available furnace parameters after the last

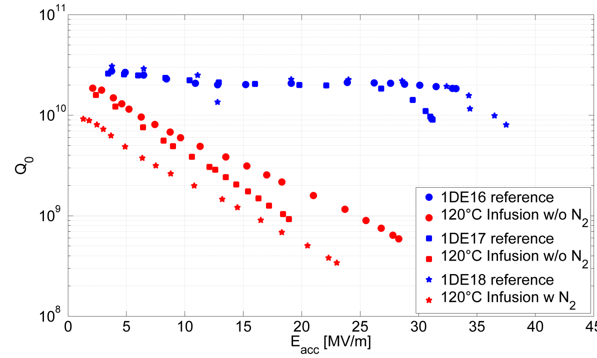

Figure 3: Quality factor versus accelerating field of three cavities. Reference measurements are blue, measurements after the treatments are red data points. 1DE18 (stars), 1DE17 (squares) and 1DE16 (circles) showed the same deterioration, regardless of the injection of nitrogen into the furnace.

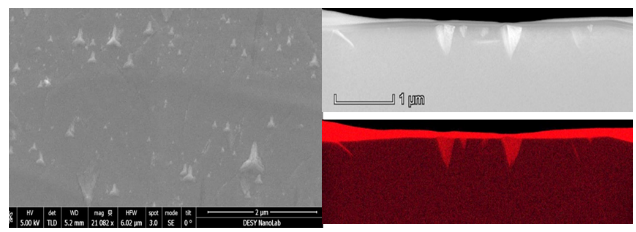

Figure 4: SEM(left) of a sample surface and a bright field TEM and TEM-EDX (top- and bottom-right) of a lamella from the same sample. Star-shaped carbon-enriched precipitates protruding from the surface are observed.

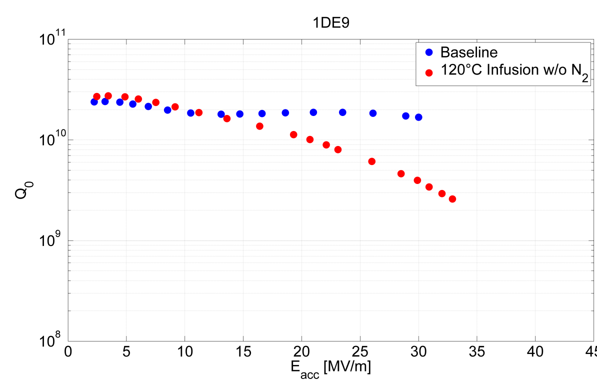

Figure 5: Quality factor versus accelerating field of 1DE9 before (blue) and after the treatment (red).

run showed a saturation in the improvements and that we achieved the best situation with given hardware. The decision for new runs with nitrogen was made. Two cavities, 1DE10 and 1DE27, were infused in two separate runs using a modified infusion recipe: In order to achieve the lowest possible pressure at 800°C and to further reduce high-mass contributions observed in the RGA, a bake at 300°C for 74 h was done in UHV before ramping up the cavity to 800°C. The rationale was to produce a further outgassing of all ma-

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terials without the creation of precipitates. The performance of the cavities is shown in Figs. 6 and 7.

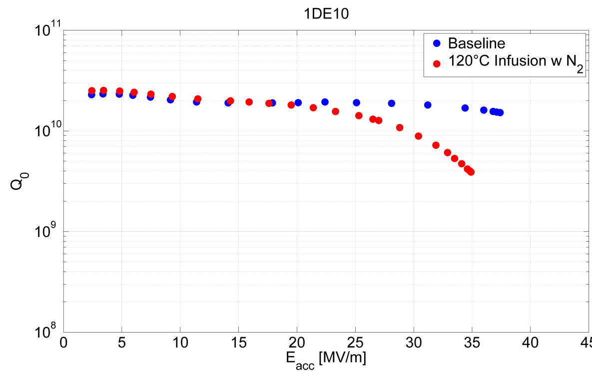

Figure 6: Quality factor versus accelerating field of 1DE10 before (blue) and after the treatment (red).

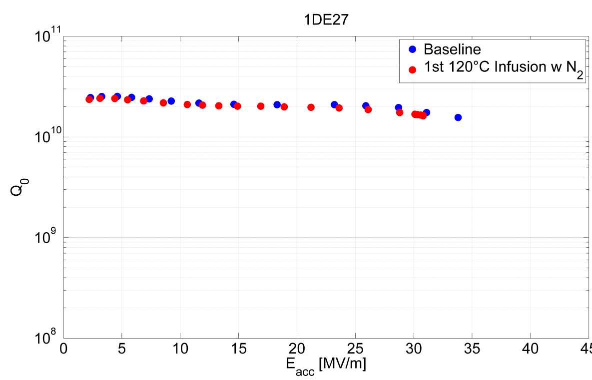

Figure 7: Quality factor versus accelerating field of 1DE27 before (blue) and after the treatment (red).

The rf performance of 1DE10 showed only a deterioration at high fields, but remained unchanged otherwise below 20 MV/m. Possible field emission - due to particle contamination during the assembly - might be the cause of this behavior. The performance of 1DE27 after the treatment is identical to the reference measurement before the treatment. During the cavity-infusion runs, the samples were baked inside the niobium box in an attempt to mimic the interior of the cavity. All samples to date have shown carbon-rich star-shaped precipitates. For the 1DE10 and 1DE27 runs, additional samples were placed on top of the box and were directly exposed to the oven atmosphere. Figure 8 shows the concentration profile obtained by TOF-SIMS of two samples which underwent the same preparation; one was baked inside the box and one was not infused.

Surprisingly, no NbN^- signal is observed, which is usually the key indicator for a successful infusion of nitrogen into niobium. The hydrogen concentration and the thickness of the oxide layer were reduced, which is expected. A strong increase of the carbon signal is in agreement with the precipitates observed with SEM and HR-TEM. Figure 9 shows a further comparison of two samples, identical except that one was placed inside the Nb box, while the other was placed on

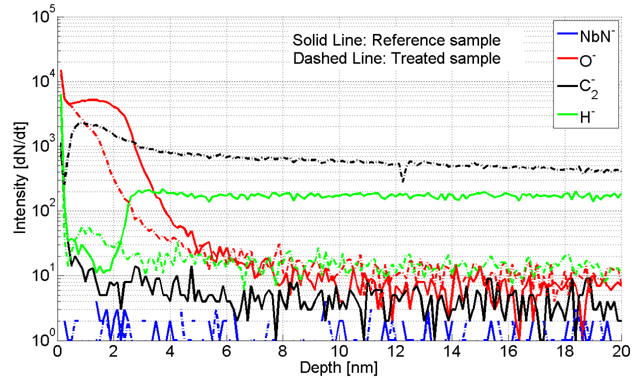

Figure 8: TOF-SIMS data of two samples, counts per second vs. depth are shown. Samples are from the same Nb sheet and were treated identically, except one was infused together with 1DE10. The solid lines are the values for the reference sample, the dashed for the treated sample.

top of it. The sample on top showed no precipitates in the SEM while they were observed on the interior sample.

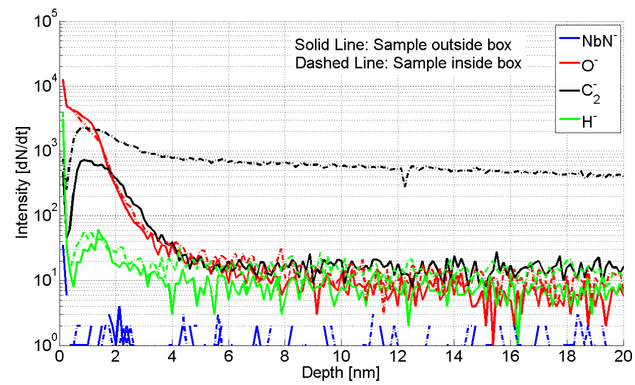

Figure 9: TOF-SIMS data of two samples, counts per second vs. depth are shown. Samples are from the same Nb sheet and were treated identically, and both were infused together with 1DE27. The solid lines are the values for the sample placed outside the box, the dashed lines for the sample placed inside.

Again, no NbN^- signal is observed in either sample. The oxide layer and the hydrogen concentration are identical except that the sample baked inside the Nb box shows a higher C concentration. This suggests that the conditions within the Nb box strongly influence the formation of the precipitates.

Cavity Cut-outs

In order to test the assumption that the samples inside the box mimic the cavity surface, 1DE16 was cut. A thorough investigation was performed to obtain spatial information on rf induced losses and other regions of interests. Eight samples were cut and underwent a dedicated surface characterization program. The first step was an SEM investigation

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to check whether the same star-shaped precipitates are observed; the result is shown in Fig. 10. The same kind of

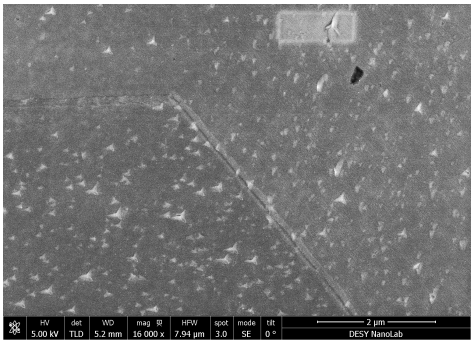

Figure 10: SEM image of a cut out from 1DE16. The same star shaped precipitates are observed as on the sample inside the Nb box.

precipitates are observed, suggesting that the Nb box as well as the caps on the cavity affect the atmosphere the Nb surface is exposed to in comparison to the open furnace volume. It gives some support to the assumption that samples do mimic the inner cavity surface. Further investigation of the cut-outs is ongoing.

In-situ Experiments

Experiments performed at DESY Nanolab [13] showed interesting results [14, 15]. Two single Nb crystals, one heated up to 2000°C for purification, the other from standard large-grain cavity grade Nb, were otherwise treated identically with 800°C for 2 hrs in UHV and then 120°C in 0.03 mbar partial pressure of nitrogen for 48 hrs during the experimental runs in the chamber. Both were investigated with SEM, but the cavity-grade material showed star-shaped precipitates similar to the samples from the cavity furnace, see Fig. 11. In addition, XPS measurements confirm the for-

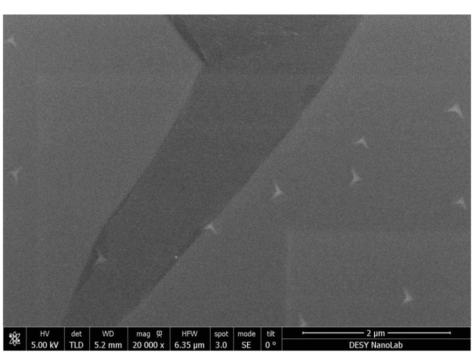

Figure 11: SEM image of a cavity-grade single-crystal sample after baking in an ultra-clean chamber. Similar star-shaped precipitates as on the cavity furnace samples can be seen. No precipitates were observed on the purified sample.

mation of a Nb-C phase present on the surface. This results directly suggest that the sample purity does play a role.

CONCLUSION

The goal of DESY infusion R&D is both reproducing the increased quality factor and high gradients demonstrated elsewhere [7, 9], and also to understand the surface evolution during this process and how this relates to the cavity performance. The three approaches - cavity treatments, in-situ sample investigation, recipe optimization - complement each other but are still in different stages of maturity. First results have suggest that the atmosphere within the cavity during the treatment is somewhat different than the open furnace atmosphere and hence hard to control or monitor. The lack of nitrogen in the TOF-SIMS measurements in all treated samples is not completely understood, but several hypothesis will be tested. A set of caps will be fabricated, to control the pressure within the cavity more accurately and make allow them to be fitted in a more reproducible way. Cavity cut-outs are under investigation to identify the origin of the deterioration of the first three cavities compared to the latter cavities, since the key parameters between all runs were too similar to cause such a difference in the performance. Clearly, not all relevant recipe parameters are yet identified.

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